INFLUENCE OF SMOKE DILUTION AND ENVIRONMENTAL CONDITIONS ON THE MEASURED LEVELS OF ETS COMPONENTS.

EFFET DE LA DILUTION ET DES FACTEURS D'ENVIRONNEMENT SUR LES CONCENTRATIONS OBSERVEES DE CERTAINS COMPOSES DE LA FUMEE AMBIANTE.

J.J. Piadé, C.J. Blake and C. Gerber
Research & Development, Philip Morris Europe, Neuchatel, Switzerland

SUMMARY

Cigarette emission yields have often been used to compute ETS exposures in indoor environments, but this approach rests on many assumptions and experimental results in this domain vary widely.
In this study, the sidestream smoke (SS) emission yields of 2 experimental cigarettes with different filter ventilations were measured in a first step using a single-cigarette total-recovery device.The SS from these cigarettes was also released into a 18 m^3 air-tight chamber, and the resulting air concentrations were measured. Eventually, the cigarettes were smoked in an office of a modern, air-conditioned building. Both human smoking and the release of SS only were studied. Different patterns of smoke generation and various room ventilation rates were tested.
The results from these sets of experiments are compared, and some implications for exposure assessment are discussed.

RESUME

On utilise souvent les rendements d'émission des cigarettes pour estimer l'exposition à la fumée ambiante dans un local. Cette approche repose sur de nombreuses suppositions et les résultats expérimentaux varient considérablement suivant les auteurs.
Dans le présent travail, on a tout d'abord mesuré les rendements émis dans le courant secondaire CS de 2 cigarettes expérimentales ayant des dilutions filtre différentes au moyen d'un collecteur individuel du CS total. Le CS fut également produit à l'intérieur d'une chambre hermétique de 18 m^3 pour mesurer les concentrations résultantes. L'expérience fut enfin répétée dans un bureau à air conditionné d'un immeuble moderne. On y a étudié à la fois le fumage humain et l'émission de CS seulement. On a testé différent taux d'échange d'air et différents profils de génération de fumée.

Les résultats de ces expériences sont comparés et les implications concernant l'estimation des expositions sont discutées.

INTRODUCTION

The investigation of the build-up and decay of environmental tobacco smoke (ETS) components has been the subject of numerous studies, both in rooms and in experimental chambers (1-9). Some mathematical models were proposed in order to describe these effects (10-12), and predictions of ETS levels were even made from minimal sets of parameters (number of air changes per hour (ACH) and room volume, number of cigarettes smoked per hour) (11). The objective of this study is to provide a comprehensive data base on the SS deliveries of experimental cigarettes, as measured under various environmental conditions. It is then to identify some pitfalls that may be encountered when trying to predict ETS exposures.

EXPERIMENTAL PROCEDURES

Experimental cigarettes

The 2 experimental cigarettes used for this study were made from virtually the same american-type blend, with no flavour or humectant addition, and designed to have filter dilutions of 16 (C20) or 46 % (C50). Kept frozen at -40 °C, they were equilibrated at 22°C and 60 % RH for 72 hours before use.

Single cigarette, total collection device

The equipment and methods used for the collection and the quantification of SS have been described previously (13). The smoking of the cigarette while enclosed in the collection device is almost identical to draft-free, open air smoking. This was assured by measuring static burn rate, mainstream smoke (MS) yields and coal temperature, and by computing air velocities inside the device (13). SS is collected as early as possible. Particulate matter is trapped on a Cambridge filter placed where the temperature of the incoming smoke is within 2 °C of ambient. Deposition inside the device is taken into account. The filter-load contents in water and nicotine are subtracted in order to approach the ETS situation more closely. Analytical procedures are very close to those employed in similar studies (14).

Experimental chamber

The equipment and the methods used for the analysis of aging SS in an air-

tight 18 m^3 stainless steel room have been described previously (4) and a comprehensive paper will be published. After smoking various numbers of cigarettes in the chamber (MS is exhausted), the air concentrations and their decays were monitored. When deriving yields per cigarette, data were extrapolated back to the middle of the smoking to compensate for decays.

Smoking sessions in an office

The office used for this study is located in a modern building with central air conditioning. It is mechanically ventilated, with 3 louvers on one wall near the perforated ceiling. It is meant for occupancy by 2 people, with a volume of 49.3 m^3. The walls are plastered and the floor carpetted. It is furnished, with 2 facing desks standing together in the center of the room. In all experiments, air mixing was assured by 2 fans located at opposite corners of the room. Smoking was performed by a smoking machine laying on a corner of one of the desks or by 2 smokers sitting at that corner. Smokers were trained to take 2-sec puffs every minute, with volumes close to 35 ml. It was verified that puff counts were virtually the same for both smokings. Sampling was performed as close as possible to where someone's head would be if he were sitting at the other desk. In some sessions, the door was partially open, while in others louvers were partially plugged.
Two types of smoke generation were performed: Pulses, where ETS decay is monitored after 2 to 4 cigarettes are smoked, and continuous generation where cigarettes are smoked at regular intervals (3-6 cig./h). In that case a steady state was reached after about 2 hours.

ETS monitoring

The procedures used for ETS monitoring are the same as those previously described (9). Aerosol particulate matter (APM) was monitored using a piezobalance (discrete measurements every 6 min.) and by filter gravimetry according to (15) (integrated over 30 min.) as a cross check on the above.

RESULTS AND DISCUSSION

Theoretical modelling of ETS concentrations

To assist in the interpretation of experimental studies on ETS performed in mechanically ventilated offices, many models have been proposed (10-12). In the present study we use a single-compartment model i.e. assume that the concentrations are uniform in the room. The release of smoke components is supposed constant during smoking and all sink rates taken as first order.

With these assumptions, and following Leaderer (10) and Repace (11), simple solutions for the mass balance differential equation can be written:

1) At equilibrium for a constant smoking rate of n cigarettes per hour, the air concentration C_{equ} of a smoke component is:

$$C_{equ} = n.E / V.k \quad \ldots [1]$$

where E is the emission per cigarette, V the room volume and k the first order constant for removal of the component. More explicitly:

$$k = m.(c_v + c_d) \quad \ldots [2]$$

where c_v is the mechanical ventilation rate, c_d is the rate of removals due to decay processes and m is a correction factor accounting for mixing dynamics (perfect mixing: m = 1)

2) During smoke concentration build-up, $C_{(t)}$, concentration corrected for the background level (so that $C_{(0)} = 0$) can be written:

$$C_{(t)} = C_{equ} (1 - e^{-kt}) \quad \ldots [3]$$

3) After smoking, when emission ceases as it reaches C_{max} and for t being the time after emission ceased, it becomes:

$$C_{(t)} = C_{max} e^{-kt} \quad \ldots [4]$$

Model testing using carbon monoxide results

Carbon monoxide is a compound for which $c_d = 0$ i.e. $k = m.c_v$ from Equ. [2]. This provides an accurate and simple way of testing the model, using data from pulse experiments with C20 cigarettes. The actual decay rate k is found by fitting the CO data in the decay phase to an exponential (equation [4]). The emission rate E can then be computed - equations [1] & [3] - from data on the smoke build-up and CO concentration peak value. Computed yields are plotted in figure 1 as a function of the decay rate k. They can be compared to those derived from chamber results:

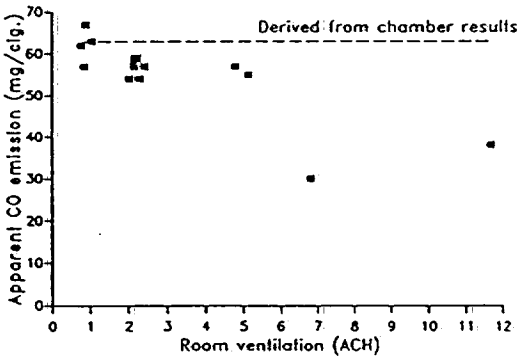

FIGURE 1: Apparent CO emission vs. room ventilation

It appears that the model is very good at low air exchange rates i.e. below 1.5 air change /hour (ACH). Results are 10 to 15 % too low thereafter, and at ACH > 6 they are only about 50% of the actual yield.

The fact that the model calls for emission yields which are lower than the actual yields outlines a bias due to the use of single compartment models: Air mixing becomes less effective as room ventilation is increased. This may result in smoke being cleared before it can be evenly distributed in

the room, where potential receivers would be. It happens here in spite of a very efficient mixing (2 fans blowing) and while the measuring spot was only 1.5 m from the source. More complex models can take this effect into account but they are not amenable to real situations (16).

Aerosol particulate matter

APM or its respirable part RSP (APM with an aerodynamic diameter < 3.5 u) have been the main target of model studies on ETS.

For APM removal, k is larger than c_v. This difference which one needs to estimate for any modelling varies with experimental conditions. It reflects losses to surfaces and evaporation (17). In air tight chambers (c_v = 0) values of 0.1 to 0.2 ACH have been reported for k (1,2) in agreement with our chamber data (0.1 ACH (4)). In this case evaporation is very limited.

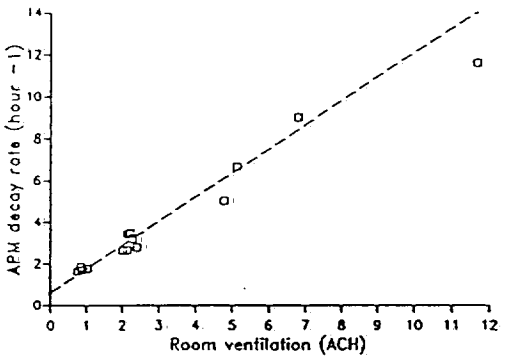

FIGURE 2: APM decay rate vs. room ventilation

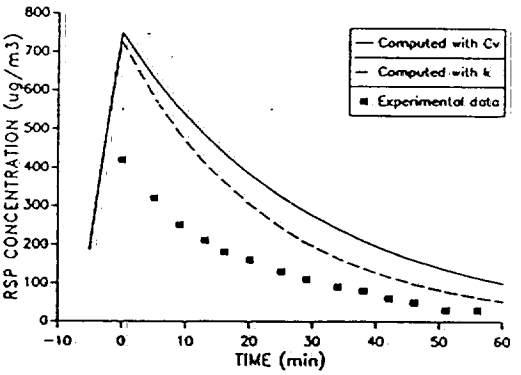

FIGURE 3: Actual and predicted APM decay

Figure 2 shows the k values computed from APM decay plotted as a function of c_v. In agreement with (10), these results show a linear relationship with a 0.6 ACH intercept. The difference between k and c_v averages 0.8 ACH with no clear influence of c_v on this value. Some models (11) assume that the ratio of k to c_v is constant. This is not supported by our data set. APM yields computed from the experimental chamber results are only 45% of the fresh SS yields, in agreement with (5, 17,18). This effect is further pronounced in ETS situations: figure 3 shows experimental data from a pulse experiment compared to the concentrations computed using either c_v (room ventilation) or k (from APM decay) as decay rates. The emission used for the calculation is that derived from chamber results. On average for all pulse experiments with less than 6 ACH, only 65% of the emission computed from the experimental room results, i.e. 30% of the actual SS yield, are actually found as ETS RSP. At very high ACH, the ratio drops further, due to clearance before mixing as discussed above. These results are summarised in table 1, as well as the computed yields of all other components studied.

TABLE 1 : Sidestream smoke yields per cigarette as obtained using the single cigarette total collector device (TCD), the experimental chamber (E.Ch.) and ETS experiments (ETS) (*)

Cigarettes	Conditions	CO (mg)	APM (mg)	NOx (ug)	NIC. (ug)	NH3 (ug)	HCN (ug)
C20	TCD	60	33.2	1760	4800	6400	n.a.
	E.Ch.	63	14.2	1850	3400	5800	270
	ETS	58	9.2	1560	390	4930	260
C50	TCD	53	30.4	1720	4500	n.a.	n.a.
	E.Ch.	53	13.7	1740	3500	6000	240
	ETS	47	8.9	1350	280	n.a.	n.a.

n.a. : not analysed (*) : Average of all experiments with room ventilation < 6 ACH

Using 24 mg as SS APM yield (value being derived from several surveys of commercial products), Repace has proposed an empirical formula to estimate the equilibrium RSP concentration in steady state situations (11):

$$C_{equ}\ (ug/m^3) = 650\ D/c_v \qquad \ldots [5]$$

D being the number of burning cigarettes per 100 m^3 of space. In our case, the C20 cigarettes used in the steady state experiments have a higher than normal APM yield of 33.2 mg so that the proposed model would be valid if:

$$C_{equ}\ (ug/m^3) = 900\ D/c_v \qquad \ldots [6]$$

Table 2 shows that equation [6] yields equilibrium concentrations that are 110 to 160 % too high, indicating that this empirical formula is grossly overestimating exposures mainly due to its failure to take into account the intrinsic difference between SS and ETS as discussed above.

TABLE 2 : Equilibrium concentrations for ETS RSP (ug/m3)

Generation rate	Comp. using [6]	Exp. data	Difference (%)
4 cig/hour	616	293	110
3 cig/hour	447	208	115
6 cig/hour	955	366	161

Nicotine

The nicotine results can be interpreted in the same way as the RSP data. Table 1 shows that even after attempting to correct for early decays, only 74% of the nicotine yield can be accounted for in the experimental chamber. It is clear that in this case the exponential extrapolation is flawed: the kinetics for wall adsorption change very fast in the early phase of the decay. For ETS, this results in even lower apparent emissions: On average for all results, the model only calls for 7.5% of the cigarette delivery.

Nitrogen oxides

In order to compare the different sets of data, the concentration of all nitrogen oxides (NOx) pooled together was taken into account. Although NOx are reactive compounds, their decay is well approximated by the exponential model in the chamber. Table 1 yet shows ETS results are still 10-20 % low.

Other ETS components

Ammonia and HCN were investigated as representative of reactive gas phase components. For practical reasons NH_3 and HCN could only be quantified in ETS in steady state experiments. Emission yields are derived from equations [1] & [2] with m = 1 and c_d set to its value in the experimental chamber. The results in table 1 show that what was observed for nicotine also takes place for NH_3 but on a smaller scale, whereas it is likely that the model could reliably be used to predict HCN concentrations in ETS.

Human smoking experiments

In fact ETS originates from both SS and exhaled MS (EMS). Some experiments where C20 cigarettes had been machine smoked were duplicated with human smoking, in order to assess the EMS contribution to ETS. On average, it was found to be 19% for CO, 46% for APM and 20% for NOx, in range with (17).

CONCLUSION

The emission yields of the SS of 2 experimental cigarettes were determined under very different environmental conditions and aging:
The fresh smoke was directly sampled using a total collection device. The smoke was also released into a room-size, air-tight chamber. Eventually SS was generated in a mechanically ventilated office and a single-compartment, exponential model was used to interpret these last results.
The fundamental difference between SS and ETS was apparent when comparing the 3 sets of experimental data: due to evaporation, the RSP concentration in ETS is only a fraction of what could be predicted on the basis of SS data. The chamber results are a better but still inadequate approximation. This is even more true for nicotine, due to its fast removal by surfaces and its complex kinetics. These biases are less pronounced but observable for other ETS components. This makes good use of ratios and markers very delicate. Most of all, it makes predictive models of ETS exposure rather unreliable, as was shown for a semi empirical formula aimed at predicting RSP levels on the basis of room parameters, SS yields and smoking rate. Even for "well-behaved" compounds like CO, biases inherent to the use of single compartment models were demonstrated: part of the emitted smoke may be evacuated before it can be distributed in the room by air drafts and diffusion, especially at higher air-change rates. Finally, the experimental cigarettes, which have very different MS yields, were found to have similar SS yields, showing the fallacy of reasoning in terms of SS / MS ratios.

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